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SID 5 Research Project Final Report

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Project identification

1. Defra Project code: WR0603

2. Project title:
   NORA: Non-Radioactive Waste Solidification Methods and Stability Assessment

3. Contractor organisation(s): University of Dundee

4. Total Defra project costs (agreed fixed price): £ 83,033

5. Project: start date .... 01 October 2006

   end date .... 31 March 2008

6. It is Defra's intention to publish this form.
Please confirm your agreement to do so.. **YES** ☒ **NO** ☐

(a) When preparing SID 5s contractors should bear in mind that Defra intends that they be made public. They should be written in a clear and concise manner and represent a full account of the research project which someone not closely associated with the project can follow.
Defra recognises that in a small minority of cases there may be information, such as intellectual property or commercially confidential data, used in or generated by the research project, which should not be disclosed. In these cases, such information should be detailed in a separate annex (not to be published) so that the SID 5 can be placed in the public domain. Where it is impossible to complete the Final Report without including references to any sensitive or confidential data, the information should be included and section (b) completed. NB: only in exceptional circumstances will Defra expect contractors to give a "No" answer.
In all cases, reasons for withholding information must be fully in line with exemptions under the Environmental Information Regulations or the Freedom of Information Act 2000.

(b) If you have answered NO, please explain why the Final report should not be released into public domain

Executive Summary

7. The executive summary must not exceed 2 sides in total of A4 and should be understandable to the intelligent non-scientist. It should cover the main objectives, methods and findings of the research, together with any other significant events and options for new work.

Executive Summary

Solidification of waste involves encapsulation in a matrix, thus isolating it from the wider environment for significant periods of time. The resulting encapsulated volume is referred to as a 'wasteform'. Wasteforms disposed of in landfill facilities may be exposed to a mobile source of water which may remove quantities of material, leading to a decline in strength. Subsequent failure is likely to lead to accelerated rates of leaching of hazardous species. Thus, there is a strong argument for including testing requirements for wasteforms which allow prediction of the extent to which physical integrity persists with time. The project described in this document was aimed at devising a test method for characterising wasteforms in terms of their likely physical stability and a system of performance criteria for either accepting or rejecting a wasteform formulation.

A literature review examined existing UK guidance relating to the solidification of waste, leach tests used by a number of countries and the range of models available for modelling leaching. The review also examined the composition of three wastes deemed potential candidates for solidification in wasteforms – incinerator fly ash, electric arc furnace dust and electroplating sludge – in terms of their chemical and mineralogical composition. Finally, data relating to the chemical nature of leachate from landfills was collected, obtained from the literature and from a UK landfill operator.

The approach taken to developing a test was to devise a means of predicting long-term degradation in strength which could be employed using data obtained from a relatively simple and short test procedure. Three leaching models were evaluated – a simple mass-transfer approach, a geochemical speciation programme (coupled with a finite-difference mass transfer model) and a 3-dimensional matrix model (again coupled with a finite-difference mass transfer model). It was necessary to divide the model evaluation into two sections, because the data from the literature used to evaluate leaching did not provide sufficient background information to provide input to the 3-dimensional matrix model. The simple mass-transfer approach yielded the best fit to the data used for evaluation.

An experimental programme was devised which was based around testing using conditions essentially identical to the NEN 7375 leach test, but incorporating measurement of loss of strength. Two wastes were studied – incinerator fly ash and a simulated electroplating sludge - whilst three wasteform matrices were used – a cement-based matrix, sintered glass and bitumen. Investigation of the effect of different specimen sizes was built into the study, along with the effect of leachant pH.

The experimental programme used to validate the use of NEN 7375 tank test leaching conditions established that leaching of bulk constituents from wasteforms seldom follow a simple diffusion-controlled

path. Instead, they display behaviour characteristic of leaching in which the leached components are becoming depleted, or in which degradation of the matrix appears to progress in an unstable manner. For this reason it has been necessary to address these different modes of leaching in the methodology for interpretation of the test results.

The need for additional components to the test procedure was also highlighted by the development of compressive strength in some cement-based specimens during the test.

Another issue to arise from the testing programme was that smaller specimens were found to produce highly variable compressive strength results when a brittle (in this case, glass) matrix was used. This has lead to the larger specimen size studied during the experimental programme to be adopted for the test methodology.

Leaching and loss of strength were also examined when the initial acidic leachant pHs were employed. One reason for taking this approach was to establish the viability of using acidic leaching conditions as a means of accelerating the degradation process. Whilst accelerated leaching was observed in many cases, some matrix / waste combinations displayed far more complex behaviour, leading to the conclusion that simple exposure to a low pH environment was provided no guarantee of accelerated degradation.

Specifically acidic conditions did accelerate leaching in cement-based wasteforms, whereas in glass-based specimens more complex behaviour was observed.

Following on from the findings of the experimental programme, methodologies have been proposed for testing and interpretation. The approach to interpretation is to use leachate conductivity measurements as a means of identifying the leaching mechanism and to use this knowledge to predict the likely decline in strength. From this, a predicted ultimate strength can be calculated which is used as a means of deciding on the adequacy of the wasteform formulation.

Testing is normally over 64 days.

Project Report to Defra

8. As a guide this report should be no longer than 20 sides of A4. This report is to provide Defra with details of the outputs of the research project for internal purposes; to meet the terms of the contract; and to allow Defra to publish details of the outputs to meet Environmental Information Regulation or Freedom of Information obligations. This short report to Defra does not preclude contractors from also seeking to publish a full, formal scientific report/paper in an appropriate scientific or other journal/publication. Indeed, Defra actively encourages such publications as part of the contract terms. The report to Defra should include:

- the scientific objectives as set out in the contract;
- the extent to which the objectives set out in the contract have been met;
- details of methods used and the results obtained, including statistical analysis (if appropriate);
- a discussion of the results and their reliability;
- the main implications of the findings;
- possible future work; and
- any action resulting from the research (e.g. IP, Knowledge Transfer).

BACKGROUND

Solidification treatment of hazardous wastes, to produce monolithic ‘wasteforms’ for landfill disposal, is an option which is becoming increasingly attractive. Solidification involves encapsulation in a matrix, thus isolating waste from the wider environment for significant periods of time. The resulting encapsulated volume is referred to as a ‘wasteform’.

At present, acceptance criteria for wasteforms for disposal in landfill facilities are that they should meet an organic carbon limit, a compressive strength test and leaching limit values for inorganic parameters of environmental significance using the NEN 7375 test.

However, the strength of almost any material is a dynamic property which will change with time. In a worst-case scenario, wasteforms may be exposed to a mobile source of water which may remove quantities of material. As material is removed, the porosity of the wasteform will increase and its strength will decline as a direct result. If failure of monolithic wasteforms occurs, a greater surface area will be exposed, leading to higher rates of leaching.

Thus, there is a strong argument for including additional requirements for wasteforms which allow prediction of the extent to which physical integrity persists with time, to ensure that scenarios such as that described above are avoided. This project aims to devise a test method for characterising wasteforms in terms of their likely physical stability and a system of performance criteria for either accepting or rejecting a wasteform ‘recipe’ based on the test results.

OBJECTIVES

The overall aim of this Project was to devise an effective method for characterising monolithic wasteforms derived from solidification treatments to establish their appropriateness in terms of long-term physical stability, thus enhancing the confidence with which such treatments can be employed. Such a method should be suitable for use as an acceptance and / or compliance testing procedure for wasteforms containing a given waste and manufactured using a given matrix material. A series of specific project objectives are listed below:

1. Conduct a review of best practice and identify and analyse models of leaching and deterioration of wasteforms containing radioactive and non-radioactive wastes.
2. Evaluate the suitability of using existing models to predict long-term performance of non-radioactive wasteforms, utilising results from the NEN 7375 tank test.
3. Develop a protocol for characterising wasteforms for predicting long-term performance in terms of physical stability.
4. Validate and refine the protocol by applying quantitative measures of goodness-of-fit to results of NEN 7375 tank tests and compressive strength measurements conducted on wasteforms containing synthetic wastes with compositions typical of UK major representatives of non-radioactive wastes suitable for solidification.
5. Develop guidance on carrying out characterisation and compliance testing to determine the physical stability of a given waste / matrix combination.

These objectives were met by the project, with the exception that the use of the test results for compliance testing was not explored, on the grounds that short testing periods were deemed inappropriate for longer term prediction of performance.

RESEARCH ACTIVITIES

To achieve the objectives the activities described below were conducted. Full details are in the Technical Report.

Objective 1 - Literature Review

A literature review was conducted into existing UK guidance relating to the solidification of waste and leach tests used by a number of countries, to achieve Objective 1. In addition, the review examined the range of models available for modelling leaching, data relating to the composition of three wastes which are potential candidates for solidification in wasteforms (incinerator fly ash, electric arc furnace dust and electroplating sludge) and data relating to the chemical nature of leachate from landfills.

The following key points emerged from the literature review:

- There is currently limited guidance on the formulation of wasteforms for non-radioactive wastes for disposal in landfill facilities.
- A range of leach tests exists but the tank and column tests are most applicable to the testing of monolithic articles such as wasteforms, and of these, the tank tests are most likely to provide leaching conditions which are most comparable to the sort of processes occurring in landfill.
- The methods used in the modelling of leaching centre around simple mass transfer models, thermodynamic dissolution models and 3-dimensional microstructural models.
- Much work has been done into predicting the mineralogical composition of a cement paste based on its initial constituents. Such an approach would be necessary if a thermodynamic dissolution model was adopted for predicting leaching. Similarly, several models exist for predicting the dissolution of glass based on its chemical composition.
- Much data exists on the composition of incinerator fly ash and electric arc furnace dust. Less data exists on electroplating sludges. However, for all materials it was evident that a broad range of compositions are possible, meaning that there is no such thing as a ‘typical’ material.

- In a number of cases, the chemical conditions measured in leachate from a number of landfill sites are relatively consistent. However, in the case of the landfill site used for the disposal of hazardous waste, which could be argued is most like the type of facility that would be used for the disposal of wasteforms, there are significant fluctuations in the chemical nature of leachate.

Objective 2 – Model Evaluation

To achieve Objective 2, a leaching model evaluation study was conducted. Three approaches to modelling were evaluated:

- A classic mass transfer model
- A 3-dimensional matrix-type model (CEMHYD3D)
- A 1-dimensional geochemical speciation model (PHREEQC)

Data from the literature was employed as the set of data used for evaluation. This data provides leachate compositions from leach testing carried out on cylindrical specimens comprising a mixture of Portland cement and incinerator fly ash tested using the NEN 7375 test. Chemical species measured in the leachate were sodium, potassium, lithium, calcium, magnesium, aluminium, silicon, iron, zinc, lead, cadmium, strontium, and barium.

In the case of the mass transfer and geochemical speciation approaches, sufficient data was provided to allow complete modelling of the leaching process. In the case of the 3-dimensional matrix model, some pertinent data was not available, meaning that evaluation in this case was deferred until experimental data was available.

Chemical species examined during evaluation were those which were present in relatively large quantities in the test specimen and, therefore, liable to have a strong influence on physical integrity. When comparing models it was necessary to select chemical species common to all models, and so calcium (Ca), aluminium (Al) and silicon (Si) were used.

Measurement of goodness-of-fit of the model to the experimental data was achieved by calculating the χ^2 value for each set of experimental and predicted data:

$$\chi^2 = \sum \frac{(O-E)^2}{E}$$

where	*O*	=	the observed (experimental) value;
and	*E*	=	the expected (predicted) value.

The criteria for the selection of a model for further application throughout the rest of the project were as follows:

- the model should provide the largest number of best fits with the experimental data with respect to Ca, Al, and Si;
- in cases where two models both perform similarly in terms of best fits, the model which over-predicts leach rates for the largest number of species would be selected.

On this basis, PHREEQC was discounted immediately on the grounds that its goodness-of-fit values were consistently higher than the mass transport model, with the exception of Aluminium. Moreover, predicted total leached values were below those of the experimental data in two instances.

The 3-dimensional matrix approach to modelling was deemed to be insufficiently flexible to permit adequate prediction in the application under investigation.

Objective 3 – Protocol Development

A ‘framework’ methodology for testing loss of compressive strength of wasteform formulations resulting from leaching was devised based around the NEN 7375 test. The reason for using this technique as the basis for the new test was that its nature mimics the types of exposure a wasteform is likely to encounter in landfill, and because it is already used as a means of establishing the suitability of wasteforms in terms of the release of environmentally significant elements. The adapted methodology included compressive strength measurements and mass loss measurements as a means of establishing physical degradation. It retained the conductivity and pH measurements from the existing method.

An experimental programme was devised to explore the validity of using this method to measure the rate of physical degradation of a series of wasteform formulations. Wastes in the form of simulated electroplating sludge and incinerator ash were used in combination with cement, glass and bitumen-based matrices. Additional test variables explored were specimen size and pH. Using a series of specimen sizes not only allowed the effect of specimen size to be studied, but also explored whether a smaller specimen size would lead to more rapid degradation, allowing for better information with regards the rate of degradation to be obtained. Two acidic leachant solutions were also used in some tests to examine the effect of lower pHs on leach rates, with the possible option of using such conditions to accelerate the degradation process.

Objective 4 – Protocol Evaluation

The experimental programme established the following key points:

- None of the specimens studied during the experimental programme displayed loss of mass behaviour during leaching which corresponds to a purely diffusion-controlled mechanism. Two types of behaviour were observed: depletion, in which the rate of release declined faster than would be expected from diffusion-controlled leaching (Figure 1), and leaching which accelerates after an initial period that would appear to be diffusion-controlled (Figure 2). A purely diffusion-controlled leaching mechanism would produce a straight line on plots of the sort shown in Figures 1 and 2.

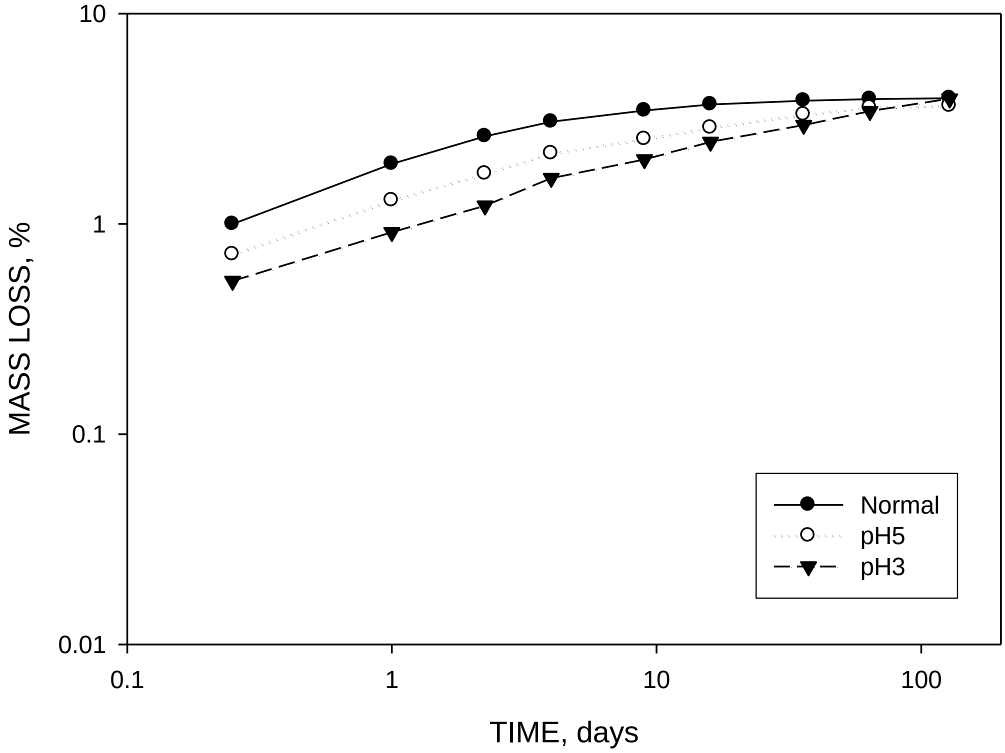

Figure 1. Cumulative mass-loss from the 50mm glass-based wasteforms containing simulated electroplating sludge.

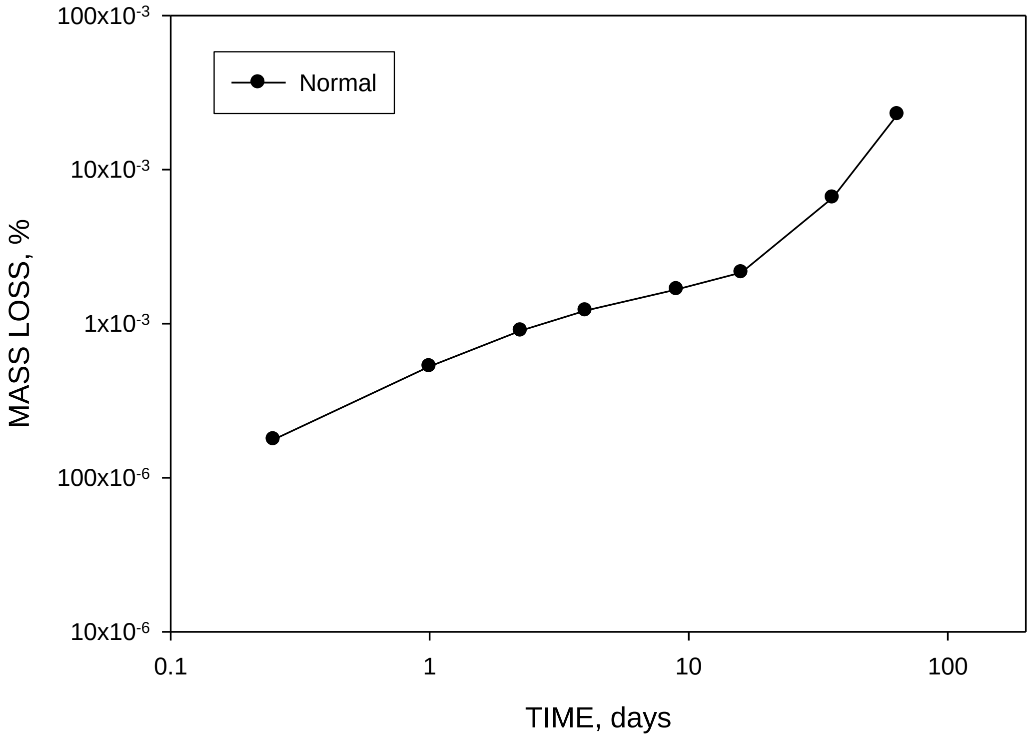

Figure 2. Cumulative mass-loss from the 75mm bitumen/sand-based wasteforms containing simulated electroplating sludge.

- In the case of the cement-based wasteforms, acidic conditions *did* accelerate leaching (Figure 3). However, in the case of the glass-based specimens, more complex behaviour was observed (Figure 1). For this reason, and because the literature review indicates that acidic conditions are not commonly encountered in landfill facilities, it is considered unwise to use acidified conditions as a means of accelerating the test procedure.

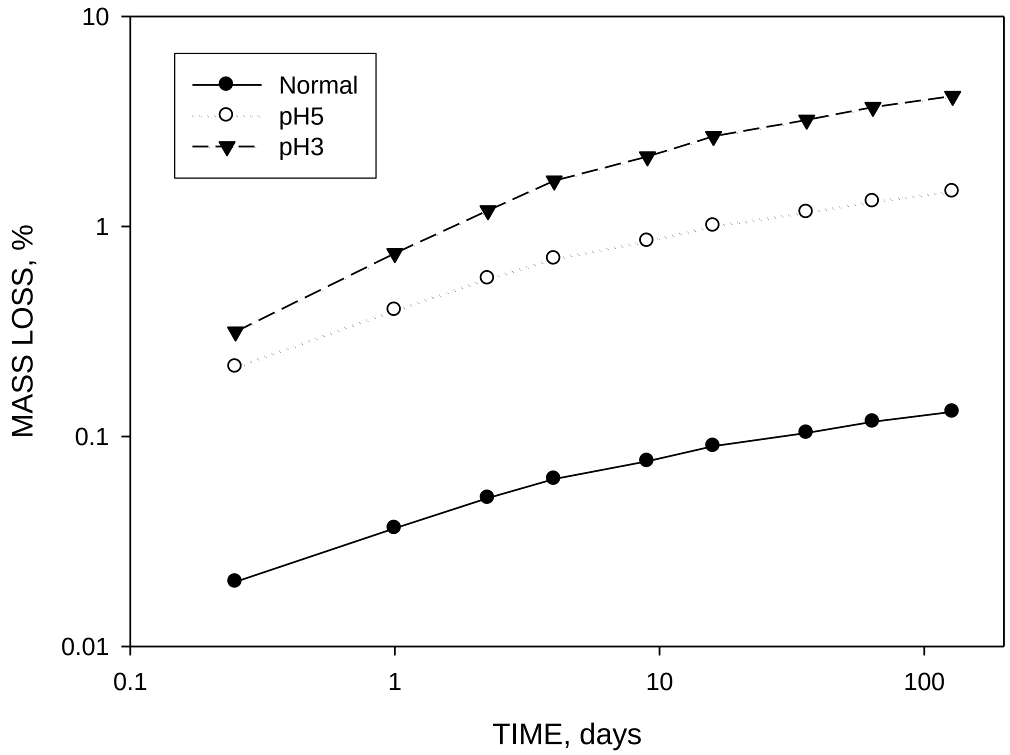

Figure 3. Cumulative mass-loss from the 50mm cement-based wasteforms containing simulated electroplating sludge.

- In certain cases, a gain in strength was observed during leach testing (Figure 4). Such behaviour required addressing by the test method.
- In the case of the cement-based specimens, compressive cube strength testing yielded adequately consistent results between specimens. However, the glass based-specimens displayed very large differences in cube strength for cube specimens less than 75mm (Figure 5). For this reason, a cube size of 75mm is recommended for testing.

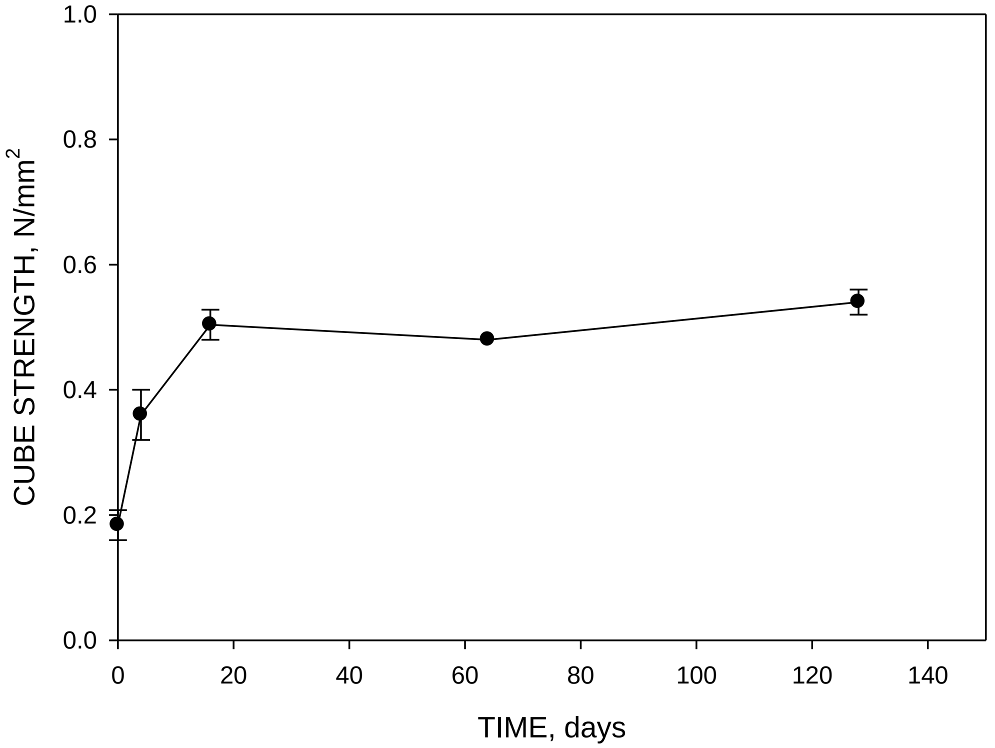

Figure 4. Compressive cube strength measurements obtained from the 25mm cement-based specimens containing simulated electroplating sludge.

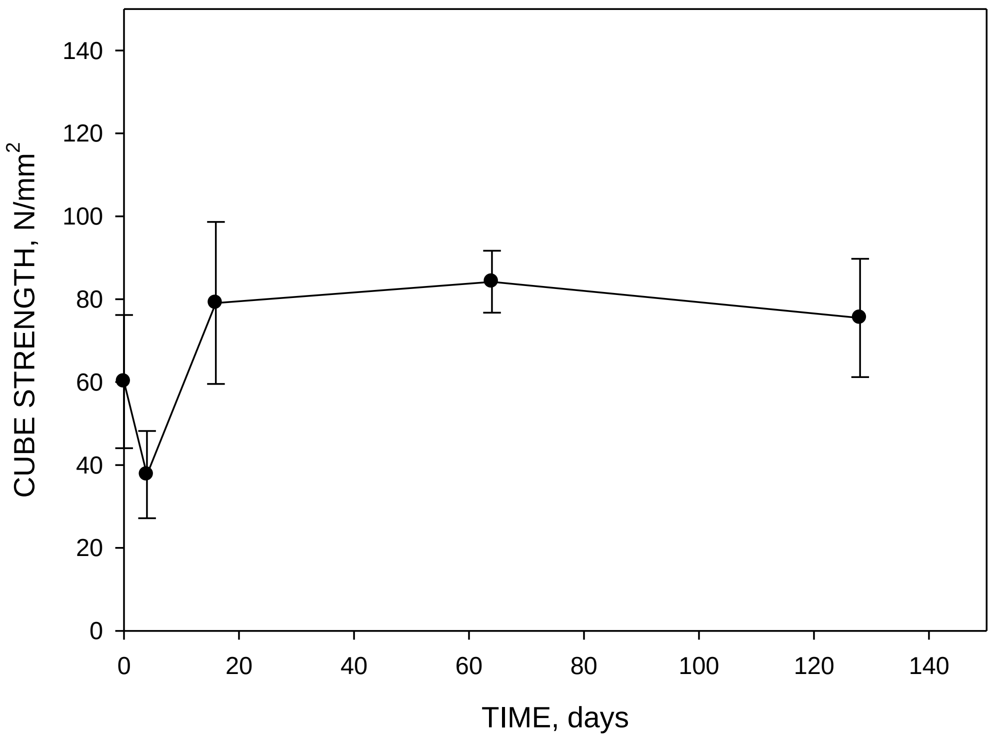

Figure 5. Compressive cube strength measurements obtained from the 25mm glass-based specimens containing simulated electroplating sludge.

- Because of the absence of purely diffusion-controlled leaching behaviour in the experimental programme, an alternative model for predicting mass-loss has been devised. The model selected was based on a hyperbolic, single rectangular, 2-parameter equation. This equation consistently under-predicts mass-loss. This problem can be overcome through the use of a factor of safety.
- The behaviour observed in which the rate of leaching increased after a period of time is more difficult to predict, since there exists uncertainty as to when and where the end-point of the leaching process is located. For this reason, no means of prediction was suggested for this leaching mechanism. Furthermore, it was proposed that the wasteform formulation should be deemed unsound for landfill disposal, on the grounds that further acceleration of leaching is possible with associated rapid loss of strength.

Objective 5 – Testing and Interpretation Methodologies

The proposed approach to the long-term prediction of degradation was based on the findings of Carde and François. This research found that the loss in strength of a cement-based article undergoing leaching was directly proportional to the ratio of the degraded cross-sectional area of the article (A_d) to its total cross-sectional area (A_t). The depth of the degraded zone is controlled by diffusion and described thus:

$$e = \sqrt{D_{app} t}$$

where e = depth of degraded zone
D_{app} = apparent (or effective) diffusion coefficient
t = time.

Hence, the thickness of the degraded zone is directly proportional to the loss of mass from the concrete, in a situation where mass loss is controlled by diffusion.

The ratio A_d/A_t for a cube is given by the equation:

$$\frac{A_d}{A_t} = \frac{l^2 - (l - 2\sqrt{D_{app} t})^2}{l^2}$$

where l = the length of one side of the cube.

Thus, the decrease in strength of a cube undergoing leaching can be described by the equation:

$$\Delta s = -b \frac{l^2 - (l - 2\sqrt{D_{app} t})^2}{l^2}$$

where Δs = the decrease in compressive strength of the cube;
and b = a constant specific to the material under investigation.

Thus,

$$s_t = s_0 - b \frac{l^2 - (l - 2\sqrt{D_{app} t})^2}{l^2}$$

where s_t = compressive strength at time t;
and s_0 = compressive strength before testing (i.e. at t=0).

Thus, by fitting a curve for a set of cubes whose strength is measured both before and after a leach test, a value for D_{app} can be obtained. Obtaining a value for D_{app} allows a prediction of loss in strength over any time period, assuming that diffusion controlled leaching persists over this time period, and the leachable constituents do not become depleted.

The previous strength decrease equation assumes that strength will continue to decline indefinitely. However, in reality the leachable constituents of a wasteform will become depleted. In a cube, the time at which depletion occurs corresponds to the time at which the depleted zones on opposite sides of the cube meet at the cube's centre. Thus, using the depletion zone depth equation above, the time at which depletion occurs in a real wasteform (t_{dep}) can be obtained:

$$t_{dep} = \frac{{l_r}^2}{4D_{app}}$$

Where a subscript ‘r’ denotes the real wasteform and its absence denotes the test specimen.

The means of establishing whether the mode of leaching is diffusion-controlled can be through examination of conductivity measurements obtained from the NEN 7375 test, since plotting this data in cumulative form on a graph with logarithmic x and y-axes should yield a straight line. It should be stressed that a cumulative conductivity plot will give an indication of the total mass of material being leached from a specimen. This means that the resulting plot would potentially be a composite of several diffusion curves (or otherwise). Nonetheless, since in most cases there will be a major constituent whose loss will have the most significant impact on strength loss, interpretation of conductivity measurements in this way is likely to be valid.

For a larger wasteform, the time taken to achieve depletion will be longer, but ultimately the same strength will be achieved. Where a state of depletion is approached during leach testing of a cube, it can be assumed that the degraded zone has reached a depth close to half the length of the cube edge. Thus, the apparent diffusion coefficient (D_{app}) can be estimated by rearranging the depletion zone depth equation and substituting 0.5*l* for *e*.

$$D_{app} = \frac{l^2}{4t}$$

Then the time at which depletion occurs in a real wasteform can be obtained on the same basis:

$$t_{dep} = \frac{l_r^{\ 2}}{4D_{app}}$$

Where depletion occurs during a test, a conductivity plot should yield a curve tending towards a horizontal line with increasing time. The approach taken to predicting the time of depletion and the extent to which leaching has occurred at this point is to use the hyperbolic, single rectangular, 2-parameter equation, as discussed in Section 5. NEN 7375 takes the approach (for leachate emissions) of estimating an upper limit for leaching which could, in theory, be adopted for this application. However, the approach taken by NEN 7375 is likely to overestimate strength loss to an extent that any wasteform would not be considered acceptable for disposal.

Using the selected equation, the depth of the degraded zone becomes:

$$e = \frac{at}{c+t}$$

where *a* and *c* are constants. Thus, the strength at time *t* is given by:

$$s_t = s_0 - b\frac{l^2 - (l - 2\frac{at}{c+t})^2}{l^2}$$

Again this equation can be fitted to strength data to obtain values for *a*, *b* and *c*.

In this case, due to the nature of the curve described by the hyperbolic equation, the time at which depletion occurs in an actual wasteform cannot be predicted. However, the strength that a wasteform will ultimately fall to can be calculated on the basis that the value of *e* tends towards *a*. Thus, the ultimate strength of the wasteform, s_{dep}, is given by:

$$s_{dep} = s_0 - b\frac{l^2 - (l - 2a)^2}{l^2}$$

Where the conductivity plot yields a curve which suggests a tendency towards the vertical, it can be assumed that this is leaching via a mechanism which suggests instability and significant deterioration of the wasteform matrix. For such results it is proposed that a wasteform formulation of this type should be deemed unsuitable for disposal.

Based on this approach a procedure for preparing specimens was proposed, along with requirements for apparatus, instrumentation, materials and reagents. The following test procedure was proposed:

Two specimens from the batch (of 10) should undergo compressive strength testing on the day that the test begins (without being exposed to leaching conditions). The test pieces should not be exposed to water prior to testing. Compressive strength testing should be carried out in accordance with BS EN 12390-3 (2002), with use of Annex B – ‘Procedure for Testing Specimens with Dimensions which are Outside the Tolerances of the Designated Sizes of EN 12390-1’. If measurement of the cubes in accordance with B.3.2.1 of this Annex identifies any dimension that is greater or less than 2 % from 75mm, the specimen should be adjusted as advised in Annex A of BS EN 12390-3, by capping with sulphur mixture. Grinding, calcium aluminate cement and sandbox methods of adjustment should not be used. Testing should be conducted using a loading rate of $0.4 \pm 0.04 N/mm^2.s$.

1. Each tank should be filled with 1265±13ml of demineralised water.

2. Unwrap the test specimen, if wrapped. The specimen must be placed such that it is in contact with the water on all sides, submerged by at least 2 cm. Seal the tank or bucket. Whilst leach test conditions are to be used for all specimens, one specimen will be used for leachate analysis. This specimen is referred to as the '*leachate analysis specimen*' below. The tank containing this specimen should be clearly marked 'Analysis'.

3. After six hours of exposure, the leachate analysis specimen should be removed from the tank and the conductivity of the leachate measured using the conductivity meter.

4. The specimen should then be immediately placed back in the tank. It should not be rinsed or dried in any way. In the case of the other specimens, the leachate solution should be poured away.

5. 1265±13ml of demineralised water should be poured into the leach tanks and the tanks resealed. This process should be repeated at the following points in the testing programme:

1 day
2 days and 6 hours
4 days
9 days
16 days
36 days
64 days

6. A small quantity of dilute nitric acid should be passed through the filtration apparatus, followed by a sufficient quantity of demineralised water to flush the apparatus of any contaminants from the leachate sample.

7. At test ages of 4, 16, 36 and 64 days, two of the specimens, other than the leachate analysis specimen should not be replaced in their tanks, but removed and subjected within 1 hour of removal to compressive testing.

The interpretation procedure is as follows:

1. On linear axes, plot compressive cube strength versus time. If strength declines throughout the test, then the procedures below can be used to further interpret the results of testing. If strength does not decline then the guidance in Section C should be followed.
2. The 'cumulative conductivity' of the leachate should be calculated for each leach test step using the equation:

$$\sigma_{cx} = \sigma_{0.25} + \sigma_1 + \sigma_{2.25} ..... + \sigma_x \qquad (1)$$

where σ_{cx} = 'cumulative conductivity' at a time of x days;
and σ_x = conductivity of leachate at a test time of x days.

3. The common logarithm of 'cumulative conductivity' (LOG(σ_c)) versus the common logarithm of time (LOG(t)) should be plotted on linear axes. From this plot, the nature of the leaching of the major available components of the specimen can be established. A linear plot indicates a leaching process which is diffusion driven (see Figure 6.1). To establish whether a plot is linear, linear regression analysis should be conducted on the plot. If the Pearson coefficient of correlation (R) is ≥0.999, then it can be concluded that leaching is diffusion-controlled, and the procedure in Section A should be followed.

   NOTE 1: The NEN 7375 test methodology utilises limits on slope values and standard deviations of slope values as a means of establishing whether leaching is diffusion-controlled. However, these limits are directly related to the quantity of leached species being released. In this instance, it is argued that the magnitude of the slope cannot be used to indicate the mode of leaching, since a major constituent of the wasteform matrix (which would yield a high slope value) could still be leaching via a diffusion-controlled mechanism. For this reason the Pearson correlation coefficient is used as a measure of linearity, since it allows measurement independent of the quantity of material leaching. The criteria for linearity adopted (i.e. ≥0.999) has been adopted from criteria for linearity in the calibration of instruments for chemical analysis.

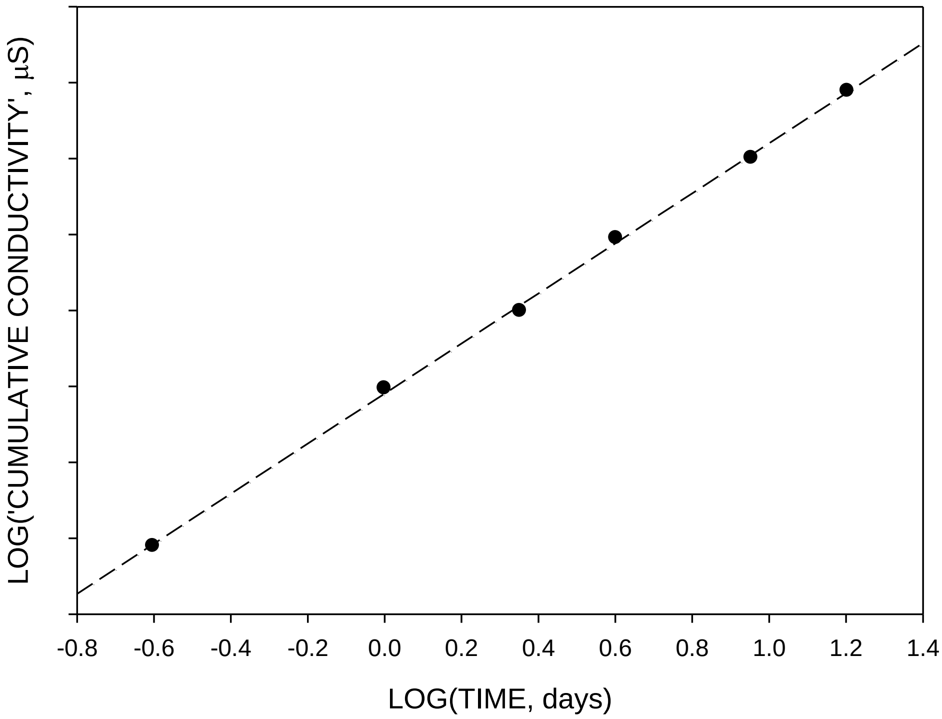

Figure 6.1. Example of a linear cumulative conductivity plot.

4. If R is <0.999, and the linear regression line at the 64-day point on the plot lies above the data plot at this age (Figure 6.2), depletion of the leached components is occurring and the procedure in Section B should be followed.

5. If R is <0.999 and the linear regression line at the 64-day point on the plot lies below the data plot at this age (Figure 6.3), it is possibly the case that the wasteform material is undergoing degradation in an unstable manner. In such cases, it is advised that the development of a wasteform formulation is revisited.

6. The test report should contain the following details:

   (a) identification of the test specimens;
   (b) age of specimens at start of leach test.
   (c) details of adjustment by capping (if appropriate);
   (d) date of tests;
   (e) compressive strengths of the specimens in Newtons per square millimetre (to the nearest 0.5 N/mm2);
   (f) unsatisfactory failure (if appropriate) and, if unsatisfactory, the closest type (as defined in BS EN 12390-2 (2000));
   (g) any deviations from the standard method of testing;
   (h) a declaration from the person technically responsible for the test that the testing was carried out in accordance with this standard, except as detailed in item g);
   (i) a common logarithm plot of cumulative conductivity versus time;
   (j) the Pearson coefficient of correlation obtained from linear regression on the plot in item (i);
   (k) a plot of cube strength versus time;
   (l) the values of constants obtained from sections A1 or B1 (if appropriate);
   (m) the predicted depleted strength of the wasteform obtained from Sections A3 or B2 (if appropriate);
   (n) a statement on the appropriateness of the wasteform formulation based on the guidance provided in Sections 5, A4 or B3 (where appropriate).

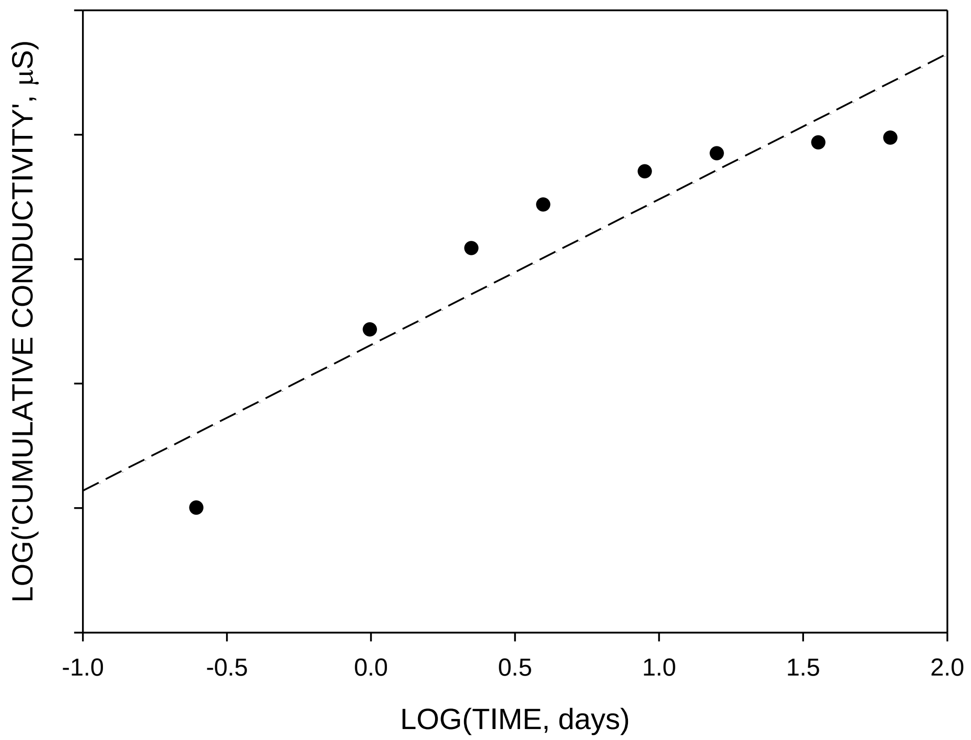

Figure 6.2. Example of a cumulative conductivity plot indicating depletion.

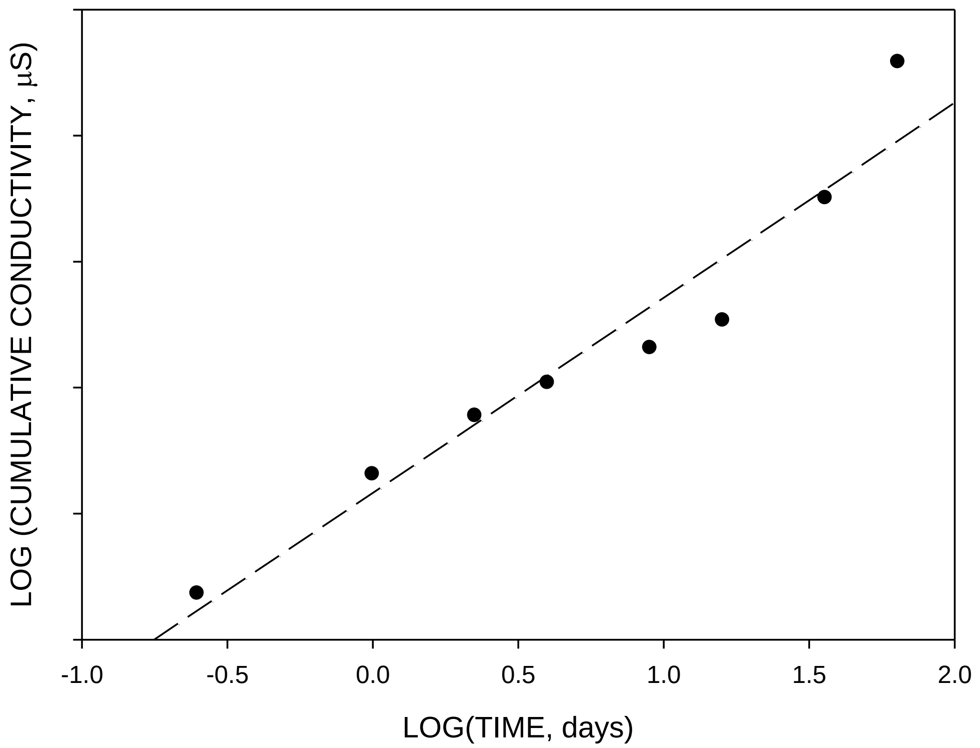

Figure 6.3. Example of a cumulative conductivity plot indicating possible unstable degradation of the matrix.

A1. A curve described by the equation:

$$s_t = s_0 - b\frac{l^2 - (l - 2\sqrt{Xt})^2}{l^2} \quad (2)$$

where
- s_t = compressive cube strength at time t (N/mm^2);
- s_0 = compressive cube strength prior to testing (N/mm^2);
- b = a constant;
- X = a constant related to the apparent diffusion coefficient of material leaching from the specimen;
- t = time (days); and
- l = length of cube edge (mm)

should be fitted to the strength measurements plotted against time to obtain values for b and X.

A2. The smallest dimension of the actual wasteform that is proposed to be used (l_r) should then be inserted into the following equation:

$$t_{dep} = \frac{l_r^{\ 2}}{4X} \qquad (3)$$

where t_{dep} = the estimated time to depletion in the actual wasteform (s).

A3. The estimated compressive strength once the wasteform has become fully depleted of leachable material (s_{dep}) is estimated using the equation:

$$s_{dep} = s_0 - b\frac{l^2 - (l - 2\sqrt{Xt_{dep}})^2}{l^2} \qquad (4)$$

This value should be expressed to one decimal place.

A4. If the predicted depleted strength (s_{dep}) is ≥4.5N/mm^2, then the wasteform formulation is appropriate for disposal purposes.

B1. A curve described by the equation:

$$s_t = s_0 - b\frac{l^2 - (l - 2\frac{at}{c+t})^2}{l^2} \qquad (2)$$

where s_t = compressive cube strength at time t (N/mm^2);
s_0 = compressive cube strength prior to testing (N/mm^2);
a = a constant;
c = a constant;
b = a constant;
t = time (days); and
l = length of cube edge (mm)

should be fitted to the strength measurements plotted against time to obtain values for *a*, *b* and *c*.

B2. The estimated compressive strength once the wasteform has become fully depleted of leachable material (s_{dep}) is estimated using the equation:

$$s_{dep} = s_0 - b\frac{l^2 - (l - 2a)^2}{l^2} \qquad (4)$$

This value should be expressed to one decimal place.

B3. If the predicted depleted strength (s_{dep}) is ≥4.5N/mm^2, then the wasteform formulation is appropriate for disposal purposes.

C1. If the compressive cube strength of the specimens increases over the test period, this indicates that the matrix used for solidification is still developing strength. If the compressive strength of the specimens remains constant, this indicates either continued strength development combined with strength loss due to leaching, or leaching at a rate which does not produce strength loss. In such cases, additional strength testing is required to establish the age at which testing should begin.

Eight cubic specimens of dimensions 75mmx75mmx75mm are required. In the case of wasteforms using a cement-based matrix, these should be prepared and cured in accordance with BS EN 12390-2 (2000). However, the method of curing in a chamber at 20°C ±2°C and a relative humidity ≥95% should be selected and the specimens should be individually wrapped in plastic film impermeable to water.

In the case of wasteforms utilising other matrices, procedures and conditions used to fabricate specimens should follow those that are to be used at full-scale.

Compressive strength measurements should be conducted on pairs of specimens at ages of 14, 28, 56 and 112 days, or until the increment in the average strength obtained from a pair of specimens between adjacent ages is less than ±10% of the last average strength value obtained. This point should then be used as the starting point of leach testing. If this point is not reached during the additional strength testing programme, it is advised that the development of a wasteform formulation is revisited.

CONCLUSIONS

Key Findings

The project conducted three main tasks. Firstly it reviewed the literature concerning the solidification of waste as wasteforms, the modelling of the leaching and degradation of wasteforms, as well as data relating to waste compositions and landfill conditions. Secondly, it has attempted to evaluate different approaches to modelling leaching. Thirdly, it has presented a proposed approach to testing (based around the Netherlands NEN 7375 tank test) and validated and modified this approach to better deal with problems and anomalies observed during an experimental programme.

The literature review examined a number of methods for leach testing and modelling of the processes occurring during leaching. It also collated and reviewed data relating to compositions of several wastes suitable for wasteform solidification and the chemical conditions encountered in landfills.

Three leaching models were evaluated – a simple mass-transfer approach, a geochemical speciation programme (coupled with a finite-difference mass transfer model) and a 3-dimensional matrix model (again coupled with a finite-difference mass transfer model). It was necessary to divide the model evaluation into two sections, because the data from the literature used to evaluate leaching did not provide sufficient background information to provide input to the 3-dimensional matrix model. The simple mass-transfer approach yielded the best fit to the data used for evaluation.

The approach taken to testing wasteform formulations was to expose specimens to NEN 7375 leach conditions, monitor the resulting decrease in strength and to use this data to predict subsequent strength loss.

The experimental programme used to validate the use of NEN 7375 tank test leaching conditions established that leaching of bulk constituents from wasteforms seldom follow a simple diffusion-controlled path. Instead, they display behaviour characteristic of leaching in which the leached components are becoming depleted, or in which degradation of the matrix appears to progress in an unstable manner. For this reason it has been necessary to address these different modes of leaching in the methodology for interpretation of the test results.

Another issue to arise from the testing programme was that smaller specimens were found to produce highly variable compressive strength results when a brittle (in this case, glass) matrix was used. This has lead to the larger specimen size studied during the experimental programme to be adopted for the test methodology.

Leaching and loss of strength were also examined when the initial acidic leachant pHs were employed. One reason for taking this approach was to establish the viability of using acidic leaching conditions as a means of accelerating the degradation process. Whilst accelerated leaching was observed in many cases, some matrix / waste combinations displayed far more complex behaviour, leading to the conclusion that simple exposure to a low pH environment was provided no guarantee of accelerated degradation.

Following on from the findings of the experimental programme, methodologies have been proposed for testing and interpretation. The approach to interpretation is to use leachate conductivity measurements as a means of identifying the leaching mechanism and to use this knowledge to predict the likely decline in strength. From this, a predicted ultimate strength can be calculated which is used as a means of deciding on the adequacy of the wasteform formulation.

Recommended Further Work

Whilst the selection of matrix and waste types has produced leaching behaviour and strength change results covering a wide range of possible outcomes using the test method, there is a strong argument for extending the range of matrix and, in particular, waste types to explore other possible modes and rates of degradation. However, the two most important issues for which further investigation is recommended are the repeatability and reproducibility of the method, and an assessment of the interpretation methodology against longer-term performance.

Repeatability (i.e. variation in measurement by a single person following the procedure) is important in this context, since it will give an indication of the adequacy of the factors of safety employed in the interpretation methodology. The reproducibility of the method (the ability of the test to be accurately replicated by anyone elsewhere) will give a general indication of the appropriateness of the proposed method, as well as providing further indication of the adequacy of the factors of safety employed. Both qualities would be best assessed in the form of a collaborative study involving multiple laboratories and multiple test specimens.

Assessing long-term performance would be most effectively achieved through field testing. In terms of assessing longer-term performance, it is unlikely that actual landfill exposure is a possibility, since recovery of wasteforms from landfill facilities would prove impractical. However, deposition of specimens for long-term exposure conditions similar to those experienced in landfill would be possible, and exposure to actual landfill leachate might also be an option. Administration and funding of such studies may also present problems, but real long-term data would be of great value in terms of validating and refining the proposed methodologies.

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9. This section should be used to record links (hypertext links where possible) or references to other published material generated by, or relating to this project.

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